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Uranium Isotopes Distribution in Soils at the Rocky Flats Plant, Colorado

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ABSTRACT

Soils east of Rocky Flats Plant (RFP) near Golden, CO, are contaminated with U as a result of past waste-storage practices, accidental release of oils laden with U, and low-level airborne emissions A spatial analysis of three U isotopes was conducted to determine the concentration and distribution pattern of U contamination Soils were sampled from 118 plots of 1 01- or 4 05-ha by compositing 25 evenly spaced samples from the top 0 64 cm Uranium-234 activity ranged from 25 9 to 92 8 Bq kg^{-1}, U-235 activity ranged from 0 1 to 25 1 Bq kg^{-1}, whereas U-238 activity ranged from 30 7 kg^{-1} to 286 Bq kg^{-1} Geostatistical techniques were used to model the spatial dependency and construct isopleth maps showing U isotope distributions Spatial correlation was not observed for U-234 This implies that U-234 is randomly distributed in the soil environment east of RFP Uranium-235 exhibited a spotty and localized concentration pattern with no clear relationship between known burial and spill sites, and the present distribution of U-235 in the soils Proposed wind-dispersal mechanisms were not consistent with the spatial distribution of U isotopes Most of the observed activities of U-234 and U-235 were well within the natural range of U isotopes in soils The lack of similarity in spatial distribution between Pu-239 + 240 and U isotopes probably resulted from the higher solubility and leachability of U isotopes compared with Pu-239 + 240 in the soil system Although U-238 exhibited a pattern of localized spatial distribution, most of its observed activity was well within the natural range of U-238 activity in soils

URANIUM CONTAMINATION of several sites at Rocky Flats Plant (RFP), near Golden, CO, is the result of accidental releases of contaminated oils from drums stored in an outside storage area, U contaminated materials buried in shallow trenches, and airborne release of U isotopes An unknown amount of U contaminated oils leaked from the drums stored at the former storage site locally known as the *903 Pad* (Fig 1) A conservative estimate based on data from Seed et al (1971) suggested that at least 31 kg of U were released at this site Approximately 1400 drums of depleted and enriched U were buried in the mound site (Fig 1) These drums were removed and shipped offsite during a cleanup operation in 1969 The oil burn pit was used in 1957 and from 1961 to 1965 to burn 1082 drums of oil contaminated with U The oil burn pit was excavated and the drums were shipped offsite Large volumes of depleted U chips and solid and oil waste were buried in the East Trenches (see Fig 1) Approximately 25 000 kg of depleted U chips and 125 000 kg of sanitary sewage sludge contaminated with U are buried in these trenches (Illsley, 1983)

The Environmental Impact Statement for RFP suggested that as much as $10^{8\,03}$ Bq measured as alpha from U-238 processing areas and $10^{8\,18}$ Bq measured as alpha from U-235 processing areas were released to the air between 1957 and 1977 Advancement in air effluent filter technology, introduced in 1970, reduced the airborne emission of U isotopes to $<10^{6\,2}$ Bq yr^{-1} (Department of Energy, 1980)

Despite the leakage of U-contaminated oils, the numerous burial sites of U-contaminated drums, and the atmospheric release of U isotopes, the magnitude and the extent of U dispersion in the soil environment around RFP has not previously been determined Moreover, there is no information regarding the U isotopic composition and the actual U concentrations in the oil that leaked from the barrels in the former storage site, the sanitary sewage sludge, and the air releases from the processing areas The impact of the U releases by RFP activities on the soil environment is also complicated by the natural abundance and variable content of U minerals in and around RFP, and the close proximity of the Schwartzwalder Uranium mine (6 4 km southwest of the plant) This mine is the largest vein-type producer of U ore in Colorado, and ranks among the six largest of this type in the USA

The chemical toxicity of U has been a primary concern in establishing control limits and procedures Uranium is chemically toxic to kidneys, and high exposure to soluble compounds can result in renal injury Insoluble U compounds are carcinogenic and tend to concentrate in the lungs when inhaled, and in the bone when ingested (Department of Energy, 1988)

The goal of this study was to provide information on the distribution of U isotopes in soils east of RFP for the purpose of risk assessment This work supports the remedial investigations and feasibility studies (RI/FS) at

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Abbreviations RFP Rocky Flats Plant RI/FS remedial investigations and feasibility studies AR activity ratio CDH Colorado Department of Health RPD relative percent difference MSE mean square error

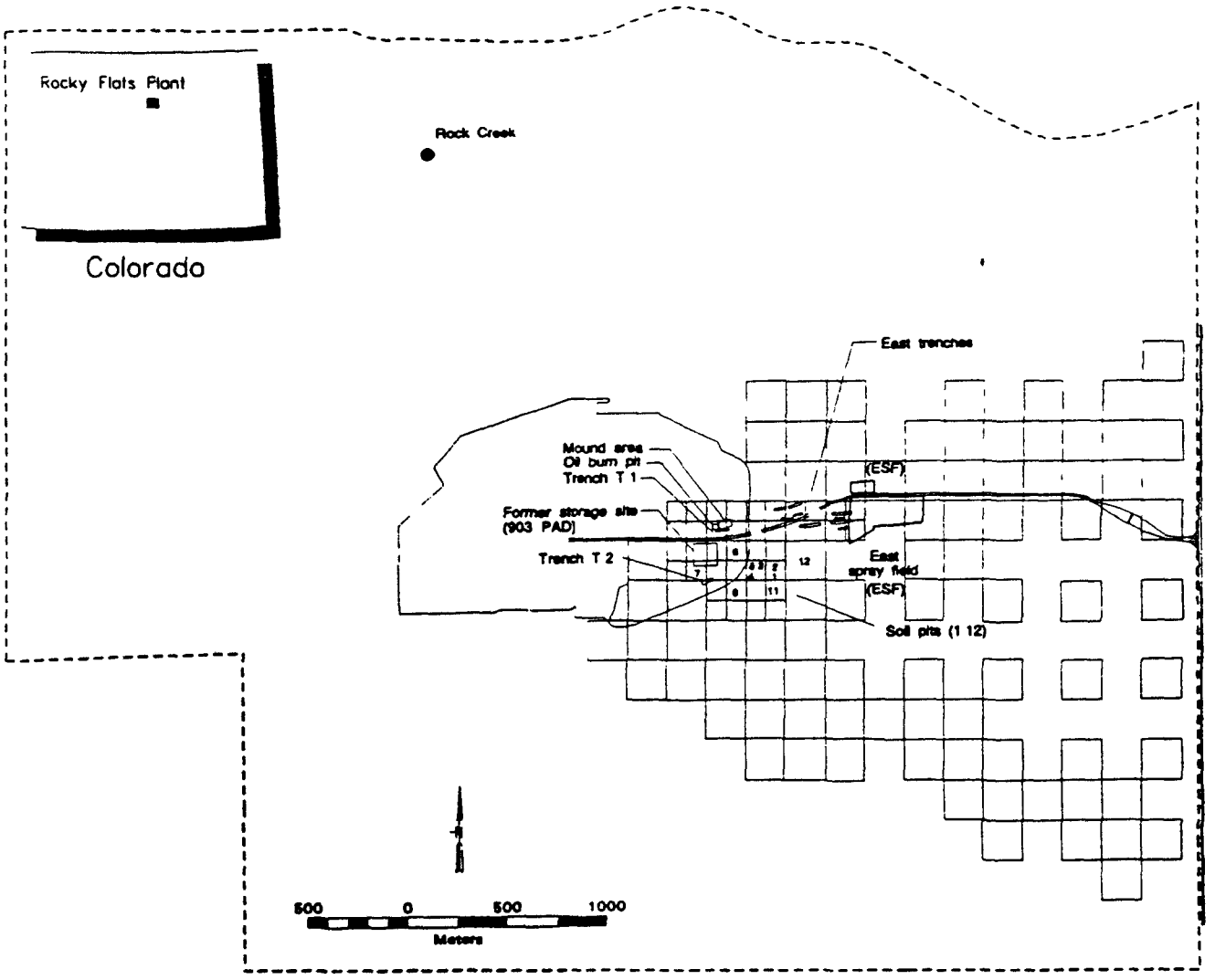

Fig 1 The location of individual hazardous substances sites containing U-contaminated drums and oils, the east spray field, and the study soil pits (1–12) The Rock Creek background study site is upwind and upslope from Rocky Flats

RFP The objectives of this study were (i) to assess the spatial distribution of U-234, U-235, and U-238 in soils around RFP, and (ii) to determine the contribution of RFP to the magnitude and extent of U isotopes in the soil environment

URANIUM GEOCHEMISTRY

The natural abundance of the three primary U isotopes are U-234, 0 005%, U-235, 0 72%, and U-238, 99 273% (Hess et al , 1985) The three primary U isotopes emit alpha (α) radiation and produce a long decay series of progeny The weapon-production facility at RFP processed enriched and depleted U. The specific activity and relative weights of the three isotopes in natural, enriched, and depleted U are summarized in Table 1

The U isotopes are commonly found in rocks such as granite, metamorphic rocks, lignites, shales, and phosphate deposits (Fairbridge, 1972) The most common minerals are uraninite (UO_2), carnotite [$K_2(UO_2)_2(VO_4)_2$], and pitchblende (variety of UO_2)

Table 1 Isotopic abundances in natural, enriched, and depleted U

Uranium isotopes	Relative weight	Specific activity†	Relative activity‡
	g kg^{-1}	Bq kg^{-1}	
Natural			
U-234	0 054	1 1E-5	6 1E-10
U-235	7 204	5 5E-7	3 9E-9
U-238	992 757	1 2E-5	1 2E-5
Enriched			
U-234	0 3	0 001	2 1E-8
U-235	29 6	2 3E-6	6 6E-8
U-238	970 1	1 1E-5	1 1E-5
Depleted			
U-234	0 005	1 1E-6	5 5E-12
U-235	2 5	1 8E-7	4 4E-10
U-238	997 5	1 2E-5	1 1E-5
Pu-239	937 9§	2 29	2 15

† Specific activity = $\lambda/(t_{1/2} \times$ atomic mass), where λ = decay constant (0 693/$t_{1/2}$ yr) Half-life of U isotopes were taken from Friedlander et al. (1981)
‡ Relative activity = relative weight × specific activity
§ Plutonium used in RFP

Table 2 Geostatistical parameters of U-234

Summary statistics (n = 118)

	Bq kg^{-1}	ln(Bq kg^{-1})		Bq kg^{-1}	ln(Bq kg^{-1})
Lowest value	25 9	3 2	75th %	51 8	3 9
25th %	37 0	3 6	Highest value	129 0	4 8
Median	44 4	3 7	Skewness	70 3	4 2
Variance	11 1	2 4	Kurtosis	247 9	5 5

Indicator variogram and model parameters

Lag class	Mean distance m	Semivariance	Pairs
1	329 9	0 35	54
2	649 4	0 29	271
3	1020 4	0 39	307
4	1402 4	0 37	395
5	1815 0	0 33	410
6	2203.3	0 33	488
7	2649 6	0 33	429
8	2973 7	0 38	362
9	3386 4	0 33	494
10	3820 6	0 37	411

The variogram for U-234 exhibited a complete nugget effect

In the soil environment, U species are found mainly in three oxidation states U(IV), U(V), and U(VI) Under most reducing conditions, U(IV) species tend to precipitate as insoluble uraninite Under most oxidizing condition U(VI) complexes are more stable than U(IV) and U(V) species An increase in oxidation state increases the mobility of U in the soil system (Langmuir, 1978)

The secular equilibrium between a radioactive parent (U-238) and a radioactive daughter (U-234) suggests that the activity ratio (AR) between these two isotopes should be 1 0 However, large deviations from this activity ratio have been reported in soils, surface water, and groundwater (Osmond and Cowart, 1976, Laul and Smith 1988) Selective leaching, diffusion, and alpha-recoil mechanisms were proposed to explain the higher mobility of U-234 compared with U-238 in different media

The historical releases of depleted and enriched U provided an excellent opportunity to use the AR concept to ascertain the impact of RFP activity on the soil environment Hence, the spatial analysis of U in soils included measurement of the three natural occurring isotopes, and analysis of their ratios

METHODS

Field Sampling

The sampling protocol for U 234 U 235 and U 238 activities in soils around RFP followed the rationale and density of sampling as described for Pu-239+240 in soils (Litaor 1995) The sampling protocol requires 25 equally spaced subsamples to be composited within 4 05- or 1 01-ha plots for U isotopes analysis The soil at each individual location was sampled with a Colorado Department of Health (CDH) sampler The sampler was designed to obtain a sample from the soil surface to a depth of 6 4 mm, and from an area 50 mm wide by 60 mm long Sampling of the top 6 4 mm of the soil may be difficult especially in stony soils The use of this technique was advocated by CDH because of the semiarid conditions in eastern Colorado that increased the potential for wind-resuspension and subsequent inhalation of soil particles containing U from the topsoil The southwest corner of each plot was located by survey and identified with an appropriately marked steel post The 25 subsamples for the composite sample were located with a hand-held compass and tape measure using the southwest corner as the starting point Eighty-four 4 05-ha plots and 34 1 01-ha plots were sampled for a total of 118 plots (Fig 1)

The most common soils in the study site were recently

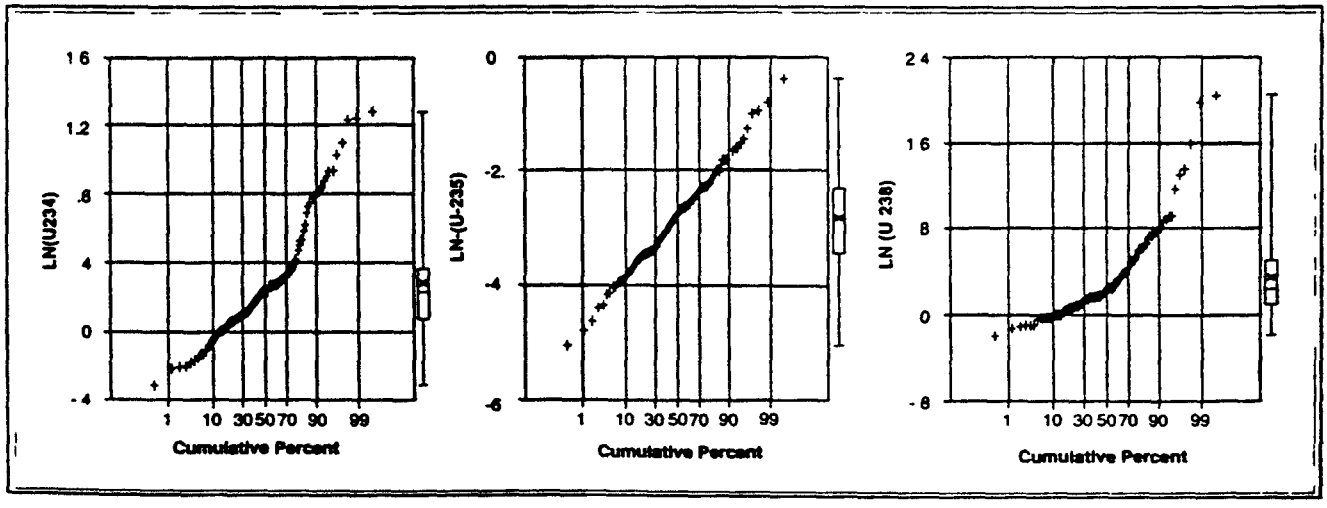

Fig 2 Cumulative distribution of U isotopes.

classified as Cumulic Haplustolls Aridic Argiustolls Pachic Argiustolls Torrertic Haplustolls Torrifluventic Haplustolls Vertic Ustochrepts Typic Ustifluvents and Aridic Haplustalfs (Litaor et al 1994)

Laboratory Analysis

The U isotopic activity in the soil samples was measured by alpha spectroscopy in several commercial laboratories These laboratories reported U-234 as U-233+234 However the natural occurring isotope U-234 is the dominant component especially in RFP where U-233 has been scarcely used The soil samples were digested using a microwave dissolution procedure (Lamothe et al , 1986 Fischer, 1986) Each sample was dissolved and digested twice with 10 mL of 8 *M* HCl The U isotopes in the samples were separated from Po by spontaneous deposition on a copper disk, from Th and Pb by anion exchange column in highly acidic solution (8 *M* HCl) and from Fe by anion exchange column in 8 *M* HNO_3 solution The U was eluted from the anion exchange column with an acidic solution (i e , pH = 1) and the sample was electroplated for alpha spectrometry

Data Quality

Duplicate soil samples were collected systematically across the study site Precision was quantified by calculating the relative percent difference (RPD) A control limit of ±35% for the RPD in laboratory duplicate analysis was recommended by the laboratory data validation guideline drafted by USEPA in 1988 There is no established criteria for the overall soil sampling and laboratory precision analysis This is because of the large variability of metals distribution in soils Indeed the RPD values for all duplicate samples ranged from 2 42 to 17 86% It should be noted, however that the highest value in each duplicate samples was used in this study

Geostatistical Approach

The first step taken in modelling the spatially correlated data was to ascertain the data distribution and minimize the spread of the data using appropriate transformations A moving-window statistical algorithm was used (Murray and Baker 1991) to assess the heteroscedasticity (unequal variances in conditional distributions) of the data The experimental semivariogram calculations and the best-fit model were developed using GS+ software (Gamma Design 1991) and GEO-EAS program (Englund and Sparks 1988) for simple and ordinary kriging and GSLIB for the indicator variogram (Deutsch and Journel 1992) Cross-validation analysis and simple and ordinary kriging computations were performed using the GEO-EAS program The universal kriging for three orders of drift was computed using a modified UVKBLK algorithm originally described by Carr (1990) The modification included universal block kriging five different types of semivariogram models and enhancements of input–output options

The mean square error (MSE) summary statistic was used to describe the bias and the spread of the error distribution (Bregt et al , 1991) The MSE from the kriging estimates was defined as

$$\mathrm{MSE} = 1/n \sum_{i=1}^{n} [z_i - z_i^*]^2 \qquad [1]$$

where z_i is the observed value and z_i^* is the estimated value The kriging technique that gave the lowest MSE the most evenly distributed error map and the smallest scatter of the observed vs the estimated plot was used for U isotopic estimation

RESULTS AND DISCUSSION

Uranium-234

Uranium-234 activity in soils around RFP ranged from 25 9 to 92 8 Bq kg^{-1}, with a median activity of 44 4 Bq kg^{-1} Most spatial estimation techniques perform better if the distribution of data is close to a normal distribution The data were transformed using a ln(Y1) function, however, there was little improvement in the measures of spread (e g , skewness, see Table 2 and Fig 2) Because the natural log transformation of the

Table 3. Geostatistical parameters of U-235

Summary statistics (n = 118)					
	Bq kg^{-1}	ln(Bq kg^{-1})		Bq kg^{-1}	ln(Bq kg^{-1})
Lowest value	0 1	−1 9	75th %	3 3	1 2
25th %	1 1	0 1	Highest value	25 1	3 2
Median	1 8	0 6	Skewness	129 1	4 8
Variance	0 3	−1.2	Kurtosis	717 4	6.5

Semivariogram & Model Parameters					
Lag class	Mean distance m	Semivariance	Pairs		
1	393 5	0 0047	101		
2	798 9	0 0065	394		
3	1328 1	0 0096	532		
4	1815 0	0 013	410		
5	2216 7	0 012	523		
6	2771.2	0 012	648		
7	3275 1	0 010	476		
	Nugget	Sill	Range, m	RSS†	r^2
Exponential	0 12	0 61	1487	0 0006	0 99

Mean square error (MSE) of the five kriging procedures	MSE
Ordinary kriging	0 72
Simple kriging	0 72
Universal kriging (k‡ = 1)	0 79
Universal kriging (k = 2)	0 66
Universal kriging (k = 3)	0 56

† RSS = reduced sum of squares.
‡ k represents order of drift.

U-234 data did little to reduce the measures of spread the nonparametric median indicator variogram discussed by Isaaks and Srivastava (1989) and Deutsch and Journel (1992) was used to assess the spatial distribution of U-234

A spatial structure was not observed with the U-234 data (Table 2), hence, a spatial estimation was not performed and an isopleth map of U-234 was not produced The lack of spatial structure suggests that U-234 is randomly distributed in the soil environment east of RFP This randomness reflects inherent irregular variation of U-234 dispersion in the soil that cannot be predicted by this sampling method Alternatively, it may represent the variability between sampling plots at distance less than that actually used, or samples collected from different populations (natural vs impacted due to RFP activity) On the basis of the available information (Table 2) the contribution of RFP to the activity of U-234 in these soils was negligible

Uranium-235

Uranium-235 activity in soils around RFP ranged from 0 1 to 25 1 Bq kg^{-1} with a median activity of 1 8 Bq kg^{-1} The data were transformed using a ln(Y1) function that reduced significantly the skewness and kurtosis (Table 3 and Fig 2) A moving-window analysis showed that the window standard deviation of the transformed data of U-235 and U-238 increased slightly with increasing window mean Thus, a universal kriging procedure

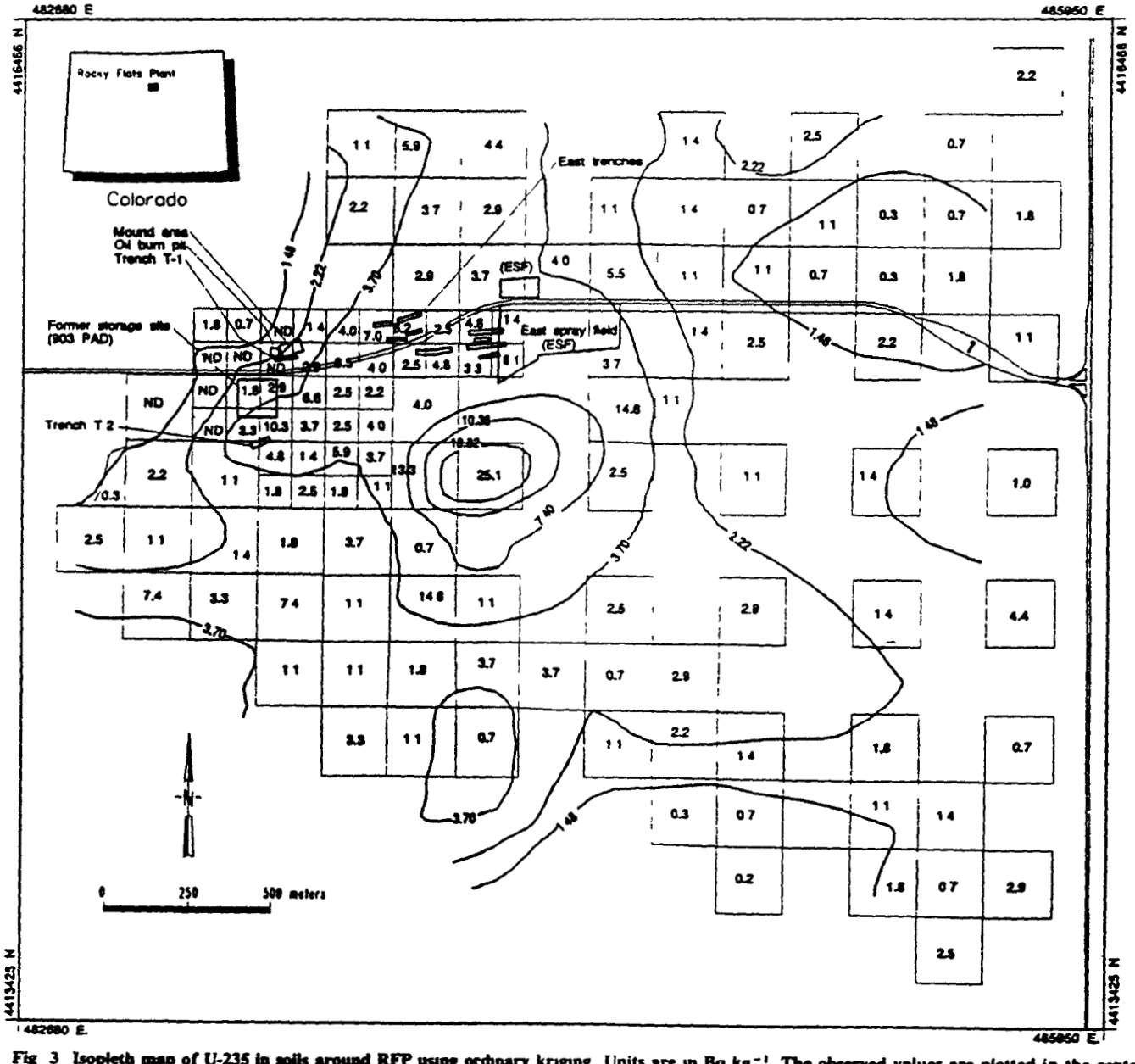

Fig 3 Isopleth map of U-235 in soils around RFP using ordinary kriging Units are in Bq kg^{-1} The observed values are plotted in the center of each 1 01- and 4 05-ha plot

that can generate good local estimation in the presence of a trend was employed

Uranium-235 was best described by an exponential model that had the lowest reduced-sum-of-squares and the highest correlation coefficient of all models tested (Table 3) Third-order universal kriging gave the lowest MSE value for U-235 data, followed by simple and ordinary kriging (Table 3) The difference in MSE between universal kriging and ordinary kriging was fairly small and, except for two pairs of observed vs estimated values, the scatter of the kriging error was the same On the basis of the MSE value, the universal kriging with cubic drift was used to estimate U-235 activity in soils around RFP

The third-order universal kriging was performed using the lognormal data for U-235 Transforming the logarithmic kriging estimates back to the original scale gave estimates of median activities Thus, to back-transform the data to the original unit of activities in terms of mean activities, the following equation was employed (Gilbert and Simpson, 1985)

$$z_i = e^{[\ln y_i + 1/2\sigma 2ki]} \quad [2]$$

where z_i is the back-transform value for a given site, y_i is the log-kriged estimate, and $\sigma^2 ki$ is the kriging variance of the log-transformed data

The resulting U-235 contour map (Fig 3) does not show the same clear west–east pattern of dispersion as those for Pu-239+240 and Am-241 (Litaor, 1995) The activity of U-235 was localized, approximately 500 m east of the industrial section of RFP (Fig 3) There was no clear relationship between the various sources of U-235 contamination considered in the present study (i e burial trenches and the leaking drums site see Fig 1) and the spatial pattern of the U-235 isopleths

The two soil plots with the highest U-235 activity (Fig 3) were probably resulted from surface flow and interflow from the east spray field (see Fig 1) The east spray field received large amount of irrigation water from a series of holding ponds that between 1952 and 1979 received laundry waste water containing actinides Surface flow and interflow from the east spray field was observed in the vicinity of these plots (J P Koffer, 1994, personal communication) Moreover, ephemeral wetland and seeps also resulted from this practice The level of U-235 in the irrigation water is unknown, however, small amounts of U-235 were observed in sediment and surface water collected from these holding ponds in 1992 (Efurd et al 1993) It was hypothesized that the small amounts of U-235 in the ponds water were reconcentrated on the soil surface of these two plots through the continuous irrigation and subsequent evapotranspiration

Uranium-238

Uranium-238 activity in soils around RFP ranged from 30 7 to 286 Bq kg^{-1}, with a median activity of 44 4 Bq kg^{-1} The data were transformed using a ln(Y1) function that only slightly reduced the skewness and kurtosis (Table 4 and Fig 2) Hence, the traditional variogram and the nonparametric median indicator variogram (Deutsch and Journel, 1992) were used to assess the spatial distribution of U-238

Uranium-238 was best described by a spherical model that had the lowest reduced sum of squares and the highest correlation coefficient (Table 4) In contrast to U-235, simple and ordinary kriging (with traditional and

Table 4 Geostatistical parameters of U-238

Summary statistics (n = 118)

	Bq kg^{-1}	ln(Bq kg^{-1})		Bq kg^{-1}	ln(Bq kg^{-1})
Lowest value	29 6	3 4	75th %	62 9	4 1
25th %	40 7	3 7	Highest value	284 9	5 6
Median	44 4	3 7	Skewness	148 0	4 9
Variance	37 0	3 6	Kurtosis	843 6	6 7

Semivariogram and model parameters

Lag class	Mean distance m	Semivariance	Pairs
1	395 4	0 030	73
2	797 9	0 034	343
3	1337 4	0 055	460
4	1823 0	0 070	350
5	2219 2	0 073	452
6	2774 4	0 071	592
7	3281 9	0 078	434
8	3755 7	0 089	487
9	4230.3	0 082	471
10	4734 1	0 068	414

	Nugget	Sill	Range m	RSS†	r^2
Spherical	0 01	0 07	2100	0 0003	0 90

Mean square error (MSE) of the five kriging procedures

	MSE
Ordinary kriging	0 10
Simple kriging	0 10
Universal kriging (k‡ = 1)	0 12
Universal kriging (k = 2)	0 10
Universal kriging (k = 3)	0 10

† RSS = reduced sum of squares.
‡ k represents order of drift.

median indicator variogram) gave the lowest MSE value for U-238, followed by third-order universal kriging (Table 4) Ordinary kriging was performed using the lognormal data for U-238 Back-transformation of the data to the original unit of activity before the map construction was performed using Eq [3] The resulting U-238 contour map (Fig 4) does not show a clear west-east dispersion pattern like those of Pu-239+240 and Am-241 (Litaor, 1995) The highest observed activities of U-238 were found around the former storage site (Fig 4), however, these values did not extend beyond the immediate vicinity of that site Plutonium-239+240 activity in the soil just east of the storage site was at least 200 times higher than U-238 activity, although at least 31 kg of U-238 were released compared with only 86 g of Pu-239+240 (see Seed et al 1971) This disparity resulted from the relative short half-life of Pu-239 ($t_{1/2}$ = 2 41 × 10^4 yr) compared with U-238 ($t_{1/2}$ = 4 47 × 10^9 yr) A comparison between the specific and relative activity of Pu-239 used in RFP and U-238 clearly demonstrates the difference between these isotopes on activity basis (see Table 1) However the complete lack of similarity in the spatial distribution across the soilscape near the former storage site could be explained by fundamental differences in solubility characteristics of these radionuclides that in turn affects their mode of dispersion in the environment Plutonium is largely insoluble in the soil environment hence upon removal of the drums during the cleanup operations the impacted area became susceptible to wind and surface erosion Conse-

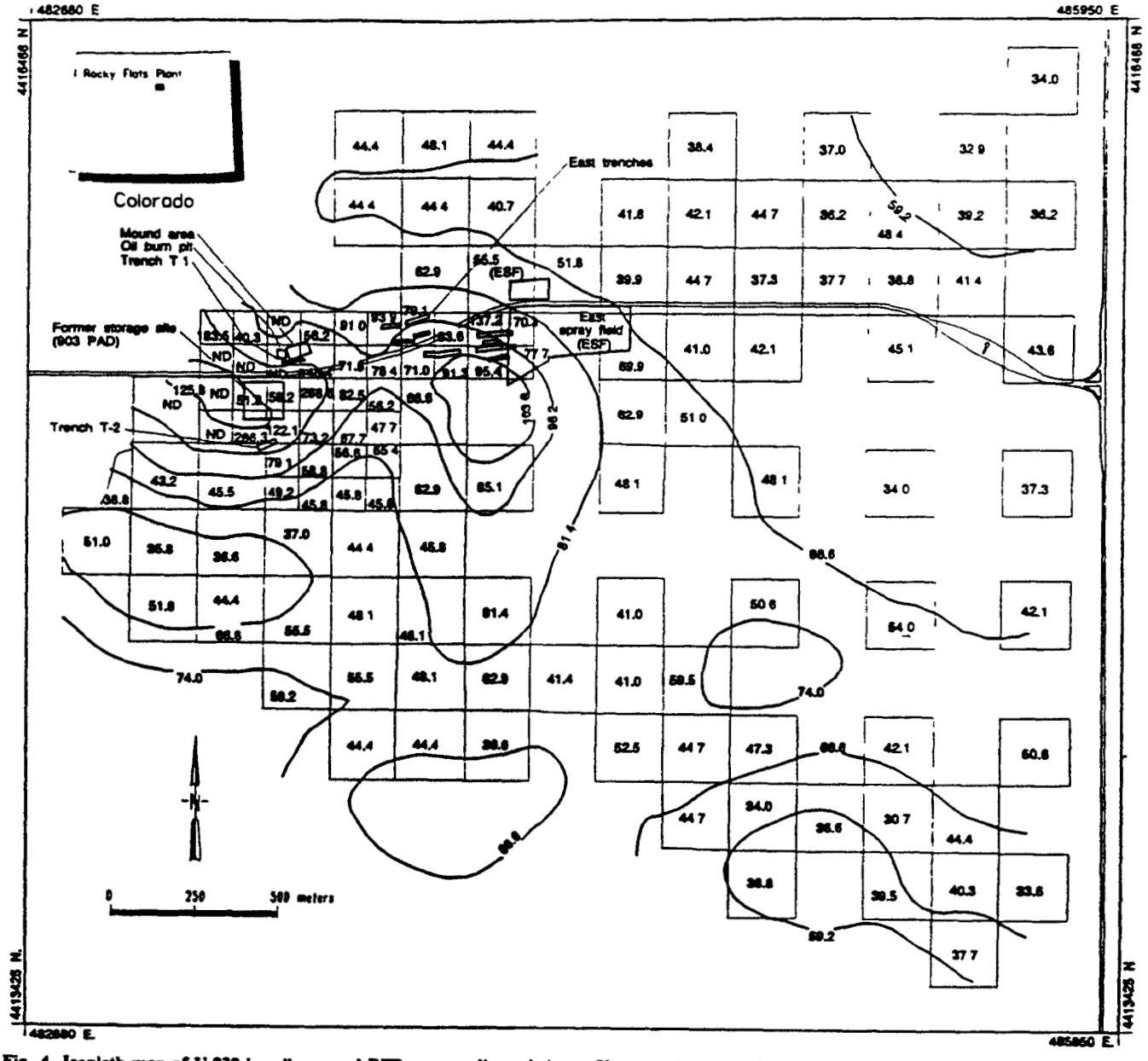

Fig 4 Isopleth map of U-238 in soils around RFP using ordinary kriging Units are in Bq kg^{-1} The observed values are plotted in the center of each 1 01- and 4.05-ha plot.

Table 5 Physical and chemical attributes of selected soils near the former storage site

Horizon†	Depth	pH	Organic matter	$CaCO_3$	CEC	Fe oxides	Sand	Silt	Clay
	cm		g kg^{-1}	k kg^{-1}	cmol kg^{-1}	mg kg^{-1}	%		
Pit-7									
A	0–2	7 0	57	1	9 9	4 850	83 0	9 5	7 3
C	2–13	6 6	9	1	3 6	5 430	90 4	4 0	5 6
2Bwb	13–39	7 3	4	1	19 4	8 040	54 0	19 5	26 5
2Btb	39–84	6 6	2	1	32 4	12 800	48 1	5 4	46 5
2BkIIb	84+	8 1	1	291	17 7	4 080	63 2	10 1	26 7
Pit-8									
A	0–3	7 7	24	1	11 6	4 950	83 1	8 3	8 6
C	3–16	8 3	13	1	17 8	5 250	84 7	6 9	8 4
2Bwb1	16–32	8 3	39	12	23 1	6 560	65 6	14 6	19 8
2Bwb2	32–50	8 2	45	242	25 4	4 330	48 5	25 5	26 0
2BmkIVb	50–105	8 1	10	369	15 7	1 880	67 3	10 2	22 6
2CkIIb	105+	8 2	1	77	11 2	5 000	81 3	5 3	13 3
Pits 1–5		*n*	KS cm h^{-1}						
A	0–18	25	13 9 ± 5 6						
Bt	30–55	19	4 0 ± 4 0						

† The original topsoil was removed by an earth grader during the cleanup operations in 1969 The entire area was covered by alluvial sand on which the A and C horizons are currently developed

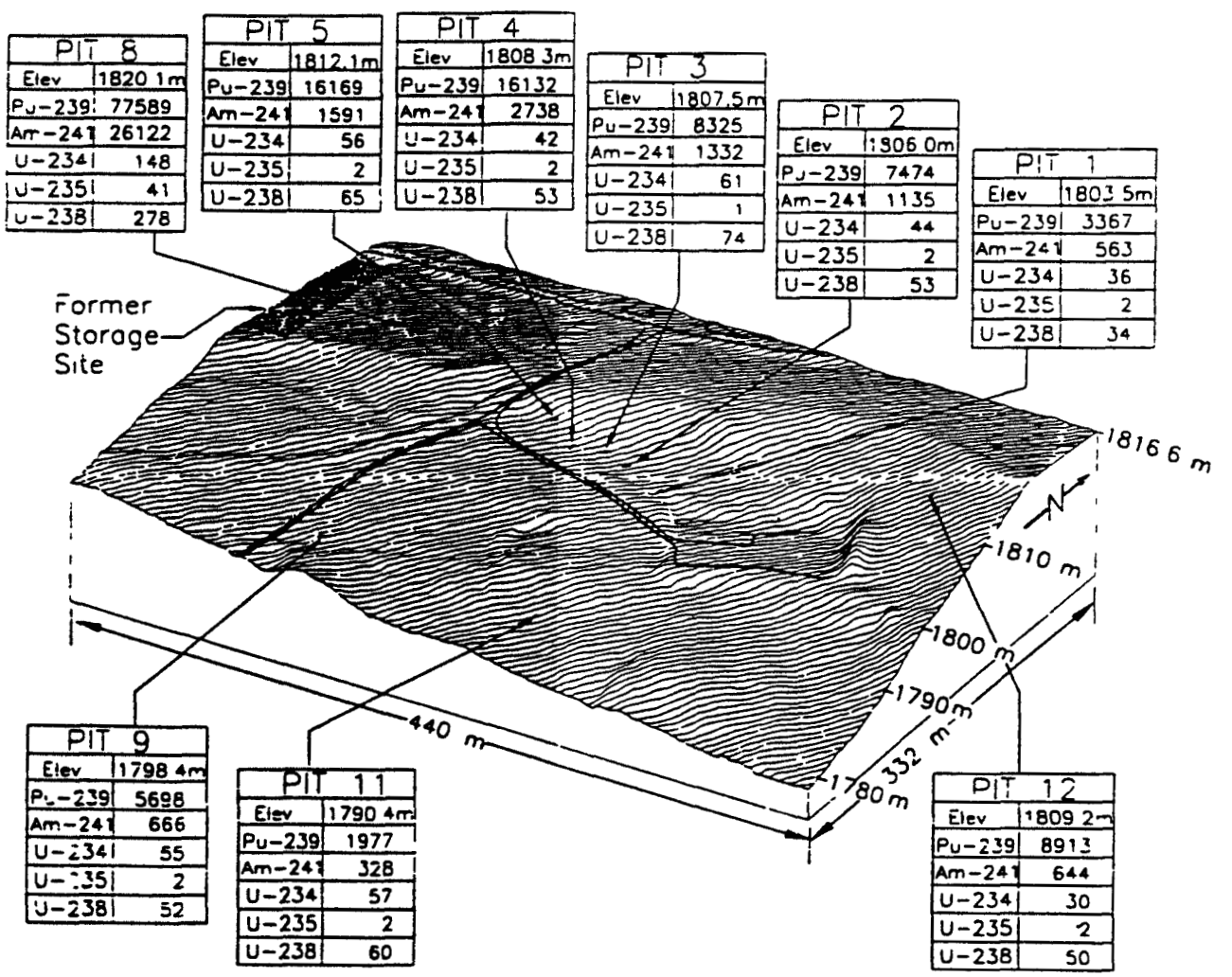

Fig 5 The activities of Pu-239/240, Am-241, and U-238 at various elevations, distances, and directions from the former storage site (units are in Bq kg^{-1})

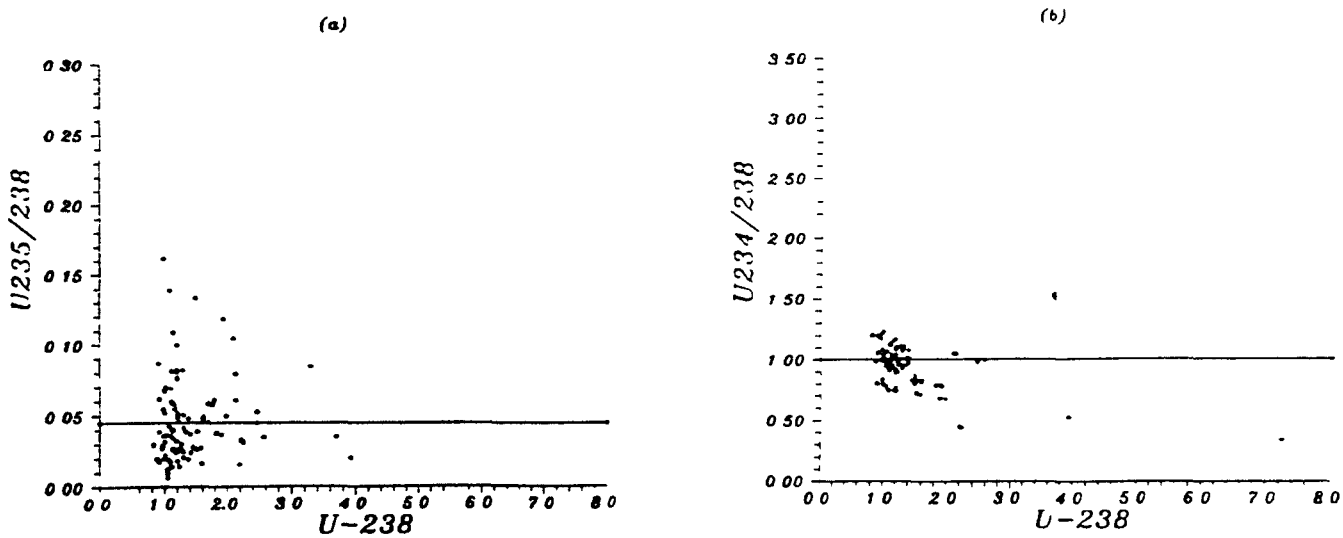

Fig 6 Scatter plot of (*a*) U-238 and U-235/238 alpha activity ratio, and (*b*) U-238 vs U 234/238 ratio

quently, Pu particles entrapped in the fine fraction of the topsoil were airlifted by winds and subsequently deposited across the soilscape east of the former storage area In contrast, under the pH and alkalinity conditions of the soil adjacent to the former storage site, U(VI) may form complex ions with carbonates and migrate downward in the soil column Indeed, Seed et al (1971) identified four *hot spots* below the asphalt cap that has been placed over the entire area of the former storage site They retrieved >31 kg of U below this cap Most of the U was concentrated above the B horizon located 15 to 40 cm below the original ground level

The U-bearing solvents that leaked from the barrels infiltrated through the A and Bw horizons, but were retarded by the 2Btb horizon due to its higher clay content (Table 5), and the 2BmkIVb horizon because of its impermeable structure Saturated hydraulic conductivity data taken from five pits at a nearby site (Pits 1–5, see Fig 1) showed that the conductivity of the A horizon was significantly greater than that of Bt horizon (Table 5) Increased clay content with depth decreased the hydraulic conductivity in the soils These flow conditions facilitated the transport of U through the surface horizons but greatly restricted the transport to greater depths (>1 m) The mobility of U was probably further restricted due to sorption of U by the sesquioxides and $CaCO_3$ minerals Because of the solubility and migratory behavior of U in the soil system, little U was entrapped in the fine particles of the topsoil Hence, wind-dispersal mechanisms did not influence the spatial distribution of U isotopes across the soilscape east of the former storage site

To test the solubility-transport mechanism, proposed for the U isotopes and compared with the wind-dispersal mechanism proposed for dispersion of Pu-239+240, eight pits were excavated east of the former storage area (see Fig 1) and analyzed for the activity of these actinides The activity of Pu-239+240 and Am-241 in the top 9 cm of the soil was highly dependent on the position and altitude across the toposequence (Fig 5) The highest activity of Pu-239+240 was observed immediately east of the former storage site, followed by the soil pit below the topographic break of the alluvial terrace on which the former storage area was located A profound decrease in Pu-239+240 and Am-241 was observed along the 150 m transect (Fig 5) This trend demonstrates that the activity of Pu-239+240 and Am-241 in the soil environment is highly dependent on elevation, distance, and direction from the former storage area This spatial distribution could only be explained by a wind dispersal mechanism On the other hand, U isotopic activities were randomly distributed in these pits with no relationship to geomorphic parameters (i e , distance, elevation, and direction from the former storage site) This finding strongly supports the hypothesis that U solubility and migration characteristics minimized U dispersal by wind across the soilscape east of the former storage area

Isotopic Ratios

The naturally occurring U-235 and U-238 are long-lived isotopes ($t_{1/2}$ = 0 71 × 10^9 yr and 4 47 × 10^9 yr, respectively) formed in a primary stellar nuclear synthesis process The minor difference in mass between U-235 and U-238 precludes significant isotopic fractionation effects by natural processes, and the U-235/U-238 abundance ratio is everywhere the same, 0 045 Indeed, Rosholt et al (1966) found that the isotopic ratio of U-235/U-238 was constant, regardless of sampling location, rock type, and degree of weathering Hence, any deviation from 0 045 attests to a soil impacted by human activity The scatter plot of U-235/U-238 ratio against U-238 activity (Fig 6a) indicates that some of the soil samples were affected by enriched U-235 However, the large analytical error associated with the determination of U-235 in soils (not shown) made the interpretation of the U-235/U-238 ratio and its spatial distribution difficult

The third naturally occurring U isotope, U-234, is relatively short-lived ($t_{1/2}$ = 248 000 yr) It is a decay

Table 6 Background activities of U isotopes in soils

Uranium isotopes	No of samples	Range	Arithmetic mean and SD
		Bq kg^{-1}	
U S avg †			
U-238	355	4 4–140	37 0 ± 30 7
Coloradot			
U-238	32	17 3–111	44 4 ± 33 6
Rock Creek‡			
U-238	21	29 6–56	41 6 ± 7 3
U-235	21	0 4–5	2 0 ± 1 3
U-234	21	28 5–54	41 9 ± 6 4

† Statistics were taken from Myrick et al (1983)
‡ Statistics compiled from a Rock Creek study west of RFP (see Fig 1) considered to be unaffected from RFP activity

product of U-238, through a sequence of intermediate, short-lived radionuclides, Th-234 and Pa-234 Hence, the abundance of U-234 in the soil system is determined by the amount of U-238 As discussed earlier, secular equilibrium predicts an activity ratio (AR) of 1 0 for the two isotopes Large deviations from this AR were observed in stream and groundwaters in many locations around the world (Osmond and Cowart 1976 Laul and Smith, 1988) The variations in U-234/U-238 ratio found in the soils under study were well within the naturally occurring range of variation (Fig 6b) Thus, the U-234/U-238 ratio cannot be used to ascertain the impact of RFP activity on the soil system

In conclusion, results of the current study indicate that the spatial distribution of U isotopes across the soilscape east of RFP is localized and does not follow the west-east plume observed for Pu-239+240 and Am-241 Uranium-234 is randomly distributed in the soil environment The observed activities of U-234 and most U-235 were well within the natural range of U isotopes in soils (Table 6) Uranium-238 showed only a localized spatial pattern and most of its observed activity lies well within the natural levels of U in soils (Table 6) These results suggest that the impact of RFP on U distribution in surficial soil is limited Hence, future remedial activities of U-isotopes in soils should focus on the directly impacted areas such as the East Trenches, the former storage site, and the few plots with elevated (i e , above background) U activities

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REFERENCES

Bregt A K A B McBratney, and M C S Wopereis 1991 Construction of isolinear maps of soil attributes with empirical confidence limits Soil Sci Soc Am J 55 14–19

Carr J R 1990 UVKRIG A Fortran-77 program for universal kriging Comp Geosci 16 211–236

Department of Energy 1980 Final environmental impact statement Rocky Flats Plant Site Golden Colorado DOE/EIS-0064 U S Dep of Energy Washington DC

Department of Energy 1988 Health physics manual of good practices for uranium facilities DOE88-013620 U S Dep of Energy Washington DC

Deutsch C V and A G Journel 1992 GSLIB Geostatistical software library and user s guide Oxford Univ Press New York

Efurd D W D J Rokop and R E Perrin 1993 Characterization of the radioactivity in surface-waters and sediments collected at the Rocky Flats facility LA-UR 93-4373 Los Alamos National Lab Los Alamos NM

Englund E and A Sparks 1988 GEO EAS geostatistical environmental assessment software user s guide USEPA Rep 600/4-88/033 USEPA Environ Monitoring Systems Lab Las Vegas NV

Fairbridge R W 1972 The encyclopedia of geochemistry and environmental sciences Vol 4A p 1215–1228 Van Nostrand Reinhold Co New York

Fischer L B 1986 Microwave dissolution of geologic materials Application to isotope dilution analysis Anal Chem 58 261–263

Friedlander G J W Kennedy E S Macias and J M Miller 1981 Nuclear and radiochemistry 3rd ed John Wiley & Sons New York

Gamma Design 1991 Geostatistics for the agronomic and biological sciences Version 2 0 Gamma Design Inc Plainwell MI

Gilbert R O and J C Simpson 1985 Kriging for estimating spatial pattern of contaminants Potential and problems Environ Monit Assess 5 113–135

Hess C T J Michel T R Horton H M Prichard and W A Coniglio 1985 The occurrence of radioactivity in public water supplies in the United States Health Phys 48 553–586

Illsley C T 1983 Environmental inventory-updated information on burial sites at Rocky Flats Internal Doc EA-321-83-240, 28 Jan 1983 Rockwell Int Golden CO

Isaaks E H , and R M Srivastava 1989 An introduction to applied geostatistics Oxford Univ Press New York

Lamothe P J T L Fries and J J Consul 1986 Evaluation of a microwave oven system for the dissolution of geologic samples Anal Chem 58 1881–1886

Langmuir D 1978 Uranium solution-mineral equilibria at low temperatures with applications to sedimentary ore deposits Geochim Cosmochim Acta 42 547–569

Laul J C and M R Smith 1988 Disequilibrium study of natural radionuclides of uranium and thorium series in cores and briny groundwaters from Palo Duro basin Texas Radioact Waste Manage Nucl Fuel Cycle 11 169–225

Litaor M I 1995 Spatial analysis of plutonium 239+240 and americium-241 in soils around Rocky Flats Colorado J Environ Qual 24 (in press)

Litaor M I M L Thompson G R Barth and P C Molzer 1994 Plutonium 239+240 and americium 241 in soils east of Rocky Flats Colorado J Environ Qual 23 1231–1239

Murray M R and D E Baker 1991 MWINDOW An interactive Fortran-77 program for calculating moving-window statistics Comp Geosci 17 423–439

Myrick T E B A Berven and F F Haywood 1983 Determination of concentrations of selected radionuclides in surface soil in the U S Health Phys 45 631–642

Osmond J K and J B Cowart 1976 The theory and uses of natural uranium isotopic variations in hydrology Atom Energy Rev 14 621–679

Rosholt J N B R Doe and M Tatsumoto 1966 Evaluation of the isotopic composition of uranium and thorium in soil profiles Geol Soc Am Bull 77 987–1004

Seed, J R K W Calkins C T Illsley, F J Miner and J B Owen 1971 Committee evaluation of Pu levels in soils within and surrounding USAEC installation at Rocky Flats Colorado RFP-INV 1 DOW Chemical Company Golden CO

10/10